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PAPER IX

Studies on the Metabolism of Fission Products
I. Studies on the Metabolism of the Radioactive Ashes Obtained from the No. 5 Fukuryu Maru

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INTRODUCTION

Some Japanese fishermen, who were working on board the No. 5 Fukuryu Maru in the Middle Pacific have suffered from radiation sickness due to the radioactive ashes produced by the hydrogen bomb test at Bikini Atoll on March lst, 1954. They complained of general malaise, anorexia, nausea, abdominal pain, diarrhea, fever, reddening and blister formation of the skin, epilation, conjunctivitis, keratitis, leukopenia etc., and these symptoms were presumed to have been caused mainly by external radiation from the radioactive ashes, which had happened to fall on the boat. However, there was a possibility that some fission products might have been absorbed from the digestive tract and respiratory tract, and that these internally deposited radioactive substances might have played some role in the development of the radiation sickness. Since the study of the metabolism of fission products was considered to be an important problem in connection with the understanding and treatment of the radiation sickness, a small amount of the radioactive ashes has been collected by us from the No. 5 Fukuryu Maru and the metabolism of fission products in animals has been studied using the radioisotopes separated from the radioactive ashes. This paper deals with the results obtained in mice by the administration of the radioactive ashes as such, hydrochloric acid extract of the ashes, precipitate at pH 7.0 and rare earth elements of the radioactive ashes.

MATERIALS AND METHODS

1) Oral administration of the radioactive ashes.

The radioactive ashes were collected from the deck of the No. 5 Fukuryu Maru by wiping the deck with wet cotton wool. About 5 grams of the cotton wool were placed in a porcelain crucible, added with a small amount of 5N NaOH, and dry

ashed for 5 hours. The ashes were pulverized with a glass rod and suspended in physiologic saline solution. After the removal of large particles, the pH of the solution was adjusted to 7.0, and the solution was diluted with distilled water so as to make the radioactivity of the solution about 20,000 counts per minute per cc. Half a cc. (or in some cases 1cc.) of this solution was administered by mouth to adult mice weighing about 15 grams, with a stomach tube attached to a syringe. From 2 to 5 mice were employed in each experiment. The animals were sacrificed 12 and 24 hours after the administration, and the liver, lungs, spleen, kidneys, blood, digestive tract (including the stomach, small and large intestines together with their contents) and bones (both femurs, tibias and fibulas, including the bone marrow) were removed. After weighing, the samples were wet ashed with perchloric acid and hydrogen peroxide, transferred into glass dishes 3 cm. in diameter, dried with an infra-red ray lamp, and the radioactivity was measured with a Geiger-Mueller counter. As a standard, the same amount of the material as the administered dose was taken in a glass dish, treated in the same way as the samples, and the radioactivity was measured under the same conditions.

2) Subcutaneous injection of the hydrochloric acid extract of the radioactive ashes.

About 4 grams of the radioactive ashes collected with cotton wool were placed in a porcelain crucible, added with a small amount of 5 N NaOH, and dry ashed for 5 hours. The ashes were extracted several times with concentrated hydrochloric acid. The hydrochloric acid was evaporated by heating and the residue was dissolved in a small volume of citric acid and hydrochloric acid. After the removal of insoluble substances, the solution was diluted with physiologic saline solution so as to make the radioactivity of the solution about 20,000 to 40,000 counts per minute per cc., and the pH of the solution was adjusted to 6.0. After sterilization at 100°C. for 30 minutes, 0.5 cc. of the solution was injected subcutaneously in the back of adult mice weighing about 15 grams. For the disinfection of the injection site tincture of iodine was used in order to prevent the mice from licking the injection site. The animals were sacrificed 4 and 24 hours after the injection, and the organs and tissues were removed, weighed and wet ashed in the same way as described above, and the radioactivity was measured with a Geiger-Mueller counter.

3) Oral administration of the precipitate at pH 7.0.

Hydrochloric acid extract of the radioactive ashes was made as described above. The hydrochloric acid was evaporated by heating, and the residue was dissolved in 6N hydrochloric acid. After the removal of insoluble substances, the solution was neutralized with ammonia. The precipitate thus formed was separated and dissolved again in 6N hydrochloric acid. This procedure was repeated three times, and then iron was removed by ether extraction for 20 hours. The ether was evaporated by heating, and the organic substances in the solution were wet ashed with perchloric acid and

hydrogen peroxide. The residual solution was neutralized with ammonia, and the precipitate thus formed was dissolved in a small volume of hydrochloric acid and citric acid. The solution was diluted with physiologic saline solution so as to make the radioactivity about 20,000 counts per minute per cc., and the pH of the solution was adjusted to 6.0. This solution was administered by mouth or subcutaneously to adult mice in the same way as described above.

4) Subcutaneous injection of rare earths.

To the precipitate at pH 7.0 described above 2.0 mg. of inert rare earths were added as carrier, and the mixture was dissolved in hydrochloric acid. The hydrochloric acid was evaporated, and the residue was dissolved again in hydrochloric acid. This procedure was repeated several times to remove silicon as much as possible. The residue was dissolved in 0.1N hydrochloric acid, and the pH of the solution was adjusted to 1.0. Rare earths were isolated from this solution using ion exchange resin, Amberlite IR-120. The method of separation will be reported elsewhere in detail. The citrate contained in the rare earth fraction was removed with perchloric acid and hydrogen peroxide. The solution was then neutralized with ammonia, and the precipitate thus obtained was dissolved in hydrochloric acid. The solution was heated to dryness and the residue was dissolved in physiologic saline solution so as to make the radioactivity 40,000 to 60,000 counts per minute per cc. The pH of the solution was adjusted to 6.0.

RESULTS

1) Oral and subcutaneous administration of the radioactive ashes (April 16th—April 26th, 1954).

The results are shown in Table 1 and Figures 1—4. In case of the oral administration the distribution of radioactivity per gram tissue was highest in the bones. The radioactivity per gram tissue of the kidneys was the second, and that of the liver was the third 12 hours following the oral administration. In case of the subcutaneous injection, the distribution of radioactivity per gram tissue was high in the kidneys, bones, liver and digestive tract in this order 4 hours following the injection. It was found that the radioactive substances were excreted not only from the kidneys but also from the digestive tract.

2) Oral administration of the precipitate at pH 7.0 (May 13th—May 16th, 1954).

The results are shown in Table 2 and Figures 5—7. Four hours following the oral administration of the precipitate at pH 7.0 the distribution of radioactivity per gram tissue was highest in the kidneys among the tissues examined except the digestive tract. As compared with the cases of the oral administration of the radioactive ashes as such the radioactivity per gram tissue of the bones was decreased, while that of the liver, lungs and spleen was increased. The radioactive

Table 1. Distribution of the radioactive ashes in the tissues of the mouse

Method of administration		Oral administration		Subcutaneous injection[3]	
Time after administration		12 hrs.	24 hrs.	4 hrs.	12 hrs.
Number of animals		3	3	2	2
Liver	p. w. o.	0.04	0.00	0.51±0.095	0.85±0.14
	p. g. t.	0.04	0.00	0.60±0.18	0.93±0.32
Spleen	p. w. o.	0.00	0.00	0.02±0.02	0.05±0.02
	p. g. t.	0.00	0.00	0.02±0.02	0.51±0.08
Lung	p. w. o.		0.07	0.02±0.02	0.15±0.02
	p. g. t.		0.10	0.02±0.02	0.57±0.26
Digestive tract[1]	p. w. o.	60.90	10.65	1.09±0.05	0.46±0.13
	p. g. t.	38.09	4.89	0.50±0.02	0.21±0.08
Blood	p. g.			7.24±3.52	0.47±0.23
Kidney	p. w. o.	0.03	0.02	0.69±0.13	1.39±0.73
	p. g. t.	0.12	0.18	2.38±0.31	4.77±3.39
Bone[2]	p. w. s.	0.34	0.35±0.03	0.34±0.42	0.45±0.01
	p. g. t.	1.12	1.75±0.15	2.17±1.31	1.45±0.32
Excreta		32.30	68.68		

p. w. o. per whole organ
p. g. per gram
p. g. t. per gram tissue
p. w. s. per whole sample

1) the stomach, small and large intestines together with their contents
2) both femurs, tibias and fibulas including the bone marrow
3) hydrochloric acid extract of the radioactive ashes
Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

Fig. 1. Distribution of the radioactive ashes in the tissues of the mouse 12 hours following oral administration (per gram).

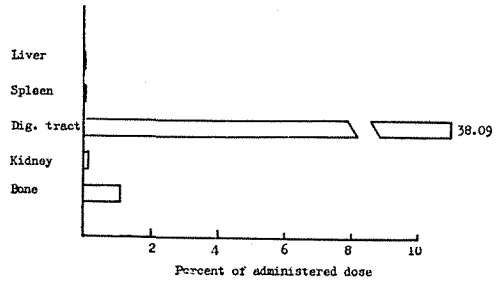


Fig. 2. Distribution of the radioactive ashes in the tissues of the mouse 24 hours following oral administration (per gram).

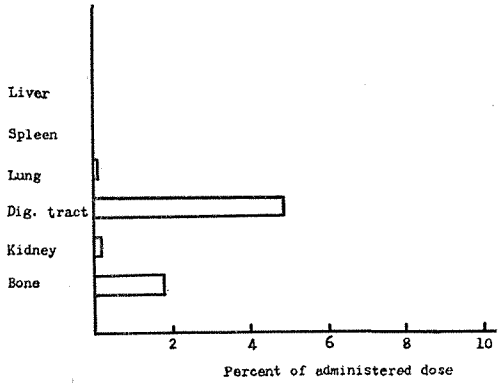


Fig. 3. Distribution of the radioactive ashes (hydrochloric acid extract) in the tissues of the mouse 4 hours following subcutaneous injection (per gram).

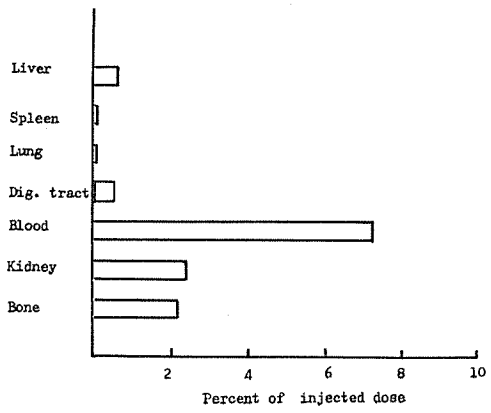


Fig. 4. Distribution of the radioactive ashes (hydrochloric acid extract) in the tissues of the mouse 12 hours following subcutaneous injection (per gram).

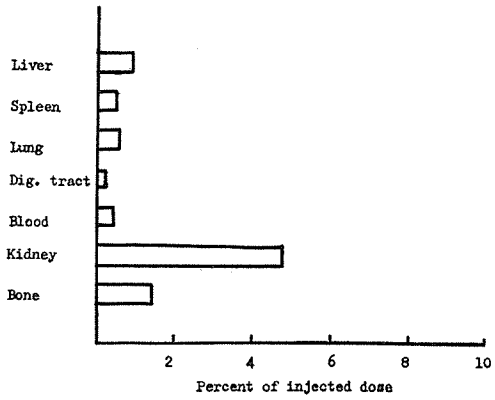


Table 2. Distribution of the precipitate at pH 7.0 in the tissues of the mouse

Method of administration		Oral administration		
Time after administration		hrs.	12 hrs.	24 hrs.
Number of animals		3	3	3
Liver	p. w. o.	0.41±0.17	0.26±0.048	0.18±0.02
	p. g. t.	0.85±0.11	0.44±0.083	0.32±0.06
Spleen	p. w. o.	0.02±0.009	0.15±0.11	0.02±0.01
	p. g. t.	0.16±0.09	2.10±1.33	0.12±0.03
Lung	p. w. o.	0.15±0.05	0.22±0.02	0.05±0.02
	p. g. t.	1.23±0.13	1.90±0.40	0.34±0.10
Digestive tract	p. w. o.	73.11±3.68	39.80±14.04	16.25±6.23
	p. g. t.	24.93±0.85	17.50±8.04	7.44±2.55
Blood	p. g.	0.42±0.26	0.19±0.07	0.05±0.02
Kidney	p. w. o.	0.43±0.13	0.06±0.01	0.11±0.04
	p. g. t.	2.26±0.65	0.31±0.06	0.75±0.24
Bone	p. w. s.	0.13±0.066	0.06±0.006	0.12±0.06
	p. g. t.	0.50±0.08	0.43±0.10	0.40±0.11

p. w. o. per whole organ
p. g. t. per gram tissue
p. g. per gram
p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

Fig. 5. Distribution of the radioactive ashes (precipitate at pH 7.0) in the tissues of the mouse 4 hours following oral administration (per gram).

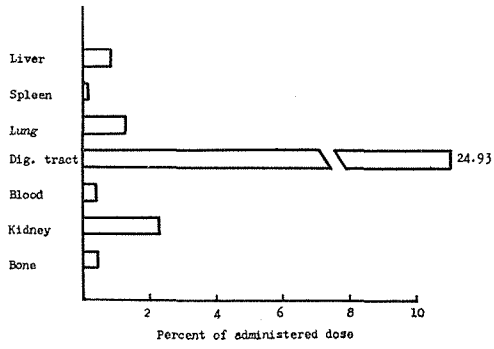


Fig. 6. Distribution of the radioactive ashes (precipitate at pH 7.0) in the tissues of the mouse 12 hours following oral administration (per gram).

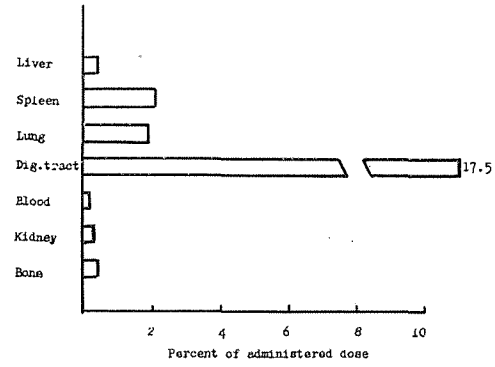


Fig. 7. Distribution of the radioactive ashes (precipitate at pH 7.0) in the tissues of the mouse 24 hours following oral administration (per gram).

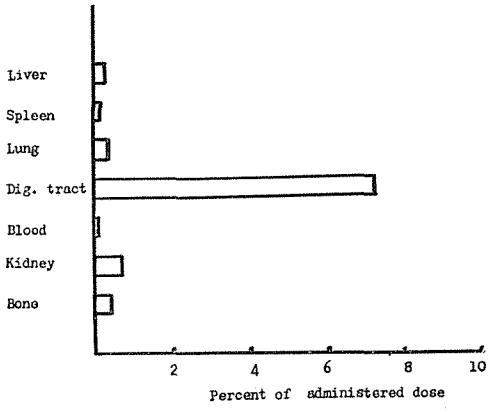
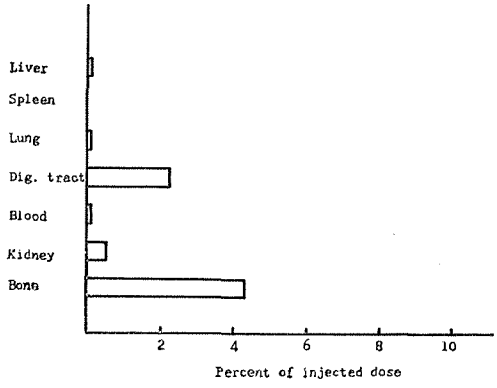


Fig. 8. Distribution of the rare earth group in the tissues of the mouse 4 hours following subcutaneous injection (per gram).



substances appeared to be excreted relatively soon. The precipitate at pH 7.0 was considered to contain rare earths (yttrium, cerium etc.), niobium, zirconium, ruthenium and rhodium. Strontium, calcium and iodine appeared to have been removed almost completely.

3) Subcutaneous injection of rare earths (May 20th—May 22nd, 1954).

The results are shown in Table 3 and Figure 8. Four hours following the subcutaneous injection of the rare earths separated from the radioactive ashes by ion exchange resin the radioactivity per gram tissue was highest in the bones. The rare earths appeared to be excreted from both the kidneys and the digestive tract.

Table 3. Distribution of the rare earth group in the tissues of the mouse

Method of administration		Subcutaneous injection
Time after administration		4 hrs.
Number of animals		5
Liver	p. w. o.	0.09±0.036
	p. g. t.	0.12±0.02
Spleen	p. w. o.	0.00±0.00
	p. g. t.	0.00±0.00
Lung	p. w. o.	0.01±0.002
	p. g. t.	0.05±0.002
Digestive tract	p. w. o.	5.14±2.20
	p. g. t.	2.29±1.37
Blood	p. g.	0.10±0.06
Kidney	p. w. o.	0.10±0.05
	p. g. t.	0.52±0.25
Bone	p. w. s.	0.89±0.29
	p. g. t.	4.35±0.82

p. w. o. per whole organ
p. g. t. per gram tissue
p. g. per gram
p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

DISCUSSION

According to the analysis made by Prof. M. Ishibashi and his co-workers[1] the composition of the radioactive ashes obtained from the No. 5 Fukuryu Maru was as follows: Ca^{45}, Sr^{89}, Y^{91}, Zr^{95}, Nb^{95}, Ru^{103}, Rh^{103m}, Ru^{106}, Rh^{106}, Te^{129}, I^{131}, Ba^{140}, La^{140}, Pr^{144}, Ce^{144}, and U^{237}.

In our animal experiments the deposition of radioisotopes was highest in the bones following the oral administration of the radioactive ashes, while in case of the oral administration of the precipitate at pH 7.0, the distribution of radioactivity was rather higher in the liver, kidneys and lungs than in the bones. These differences might be attributed to the following two facts. Firstly, in the precipitate at pH 7.0, alkaline earths, such as strontium, barium, calcium etc., were not contained in contrast with the radioactive ashes as such. Secondly, in case of the oral administration of the radioactive ashes as such, the material was dry ashed and suspended in physiologic saline solution, while in case of the oral administration of the precipitate at pH 7.0 the material was dissolved in hydrochloric acid and added with a small amount of citric acid. Judging from the results obtained in our experiments, the radioisotopes which were deposited in the bones following the oral administration of

the radioactive ashes as such, were chiefly of the alkaline earth group, and in case of the oral administration of the precipitate at pH 7.0 the radioisotopes which were deposited in the body, were chiefly heavy metals such as ruthenium, rhodium, zirconium and niobium. There was a possibility, however, that the absorption of these heavy metals was enhanced by the addition of citric acid.

In case of the subcutaneous injection of the hydrochloric acid extract of the radioactive ashes the radioactivity per gram tissue was high in the kidneys, bones, liver and digestive tract in this order, while in case of the subcutaneous injection of the rare earth group the radioactivity per gram tissue of the bones was the highest, followed by that of the digestive tract, kidneys and liver in this order. Judging from these results the rare earth group appeared to be deposited chiefly in the bones and excreted from the kidneys and digestive tract. Since the radioelements of the alkaline earth group, such as strontium, calcium and barium are said to be deposited chiefly in the bones in the same way as rare earths, the main sites of deposition of the heavy metals following the subcutaneous injection of the hydrochloric acid extract of the radioactive ashes appeared to be the liver, lungs and spleen. The heavy metals appeared to be excreted mainly from the kidneys. There was also a possibility that the heavy metals in colloidal form were taken up by the reticuloendothelial system in the spleen, liver and other organs.

SUMMARY

1) The metabolism of the radioisotopes contained in the radioactive ashes collected from the No. 5 Fukuryu Maru has been studied in adult mice.

2) When the radioactive ashes were administered by mouth, the radioisotopes which were chiefly absorbed, were alkaline earths, and these elements were deposited mainly in the bones.

3) When, after the removal of the alkaline earths, the radioisotopes contained in the radioactive ashes were administered by mouth in the form of the chloride or citrate, the radioisotopes which were chiefly absorbed, were the heavy metals such as radioruthenium and radiorhodium, and the main sites of deposition of these elements were the kidneys, liver, lungs and spleen. The heavy metals appeared to be excreted chiefly from the kidneys.

4) The rare earths were deposited mainly in the bones following subcutaneous injection, and excreted from the kidneys and digestive tract.

5) The hydrochloric acid extract of the radioactive ashes, when administered subcutaneously, was deposited in the bones, kidneys, liver, lungs and spleen, and excreted from the kidneys and digestive tract.

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1) Ishibashi, M. *et al.* : Radiochemical analysis of the Bikini Ashes. Paper VII., This issue, 1954.